

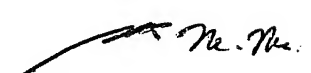
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No. : 10/541,471
Applicants : John Sydney Robinson et al
Filed : 02/15/2006
Title : Process for the Production of Plate-like Alumina Particles
Group Art Unit : 1793
Examiner : James Fiorito
Confirmation No. : 7786

DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

1. I, Michihito Muroi, resident in Nedlands, Western Australia, am familiar with the subject matter of the above-identified application and the invention described therein ("**the present invention**"). I am not an inventor of the present invention. I have worked on inorganic materials for more than 20 years both in industry and academia.
2. I am currently employed as a Senior Product Development Leader by Antaria Limited. I was employed as a Research Scientist / Senior Research Scientist by Antaria Limited (formerly Advanced Nanotechnology Limited) in the period 2002-2008. From 1995 to 2001, I held the position of Research Fellow or Research Associate at the University of Western Australia. From 1985 to 1990, I was employed as a Research Engineering by Fuji Electric Corporate Research and Development Ltd in Japan.
3. I hold a Bachelor of Liberal Arts degree (Pure and Applied Sciences) from the University of Tokyo, a Master of Engineering degree (Energy Sciences) from Tokyo Institute of Technology and a PhD with distinction (Physics) from the University of Western Australia.

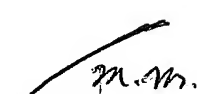
 M. Muroi

4. I am fluent in Japanese, having been born and raised in Japan. I moved to Australia in 1990. I am thus able to read and understand documents published in the Japanese language and in the English language in the art of inorganic materials processing.
5. I have been provided with a copy of JP2001058818 (hereinafter referred to as "JP'818") which I have read and understood in full. JP'818 discloses a process in which gamma-alumina is heated with a flux, sodium sulfate, to form alpha-alumina plate-like particles. An example in JP'818, however, clearly shows that single-phase alpha-alumina is formed only after heating to 1000°C; at 800°C no alpha-alumina is formed. The flux used in JP'818 is sodium sulphate which (according to Wikipedia) has a melting point of 884°C. JP'818 teaches that heat treatment must be conducted above 900°C, ie above the melting point of the diluent in order to form **plate-like alpha alumina**.
6. In contrast, the present invention is directed towards the formation of plate-like alpha alumina particles at a temperature **below** the melting point of the diluent. This difference is very significant in that a process that can be done below the melting point of the diluent is both cheaper and easier to operate which makes the present invention highly advantageous compared to the process described in JP'818. I consider the JP'818 document to be irrelevant to the present invention.
7. I have been provided with a copy of US Patent 6,521,203 (hereinafter referred to as "the Mohri reference") which I have read and understood. Mohri discloses a method for the synthesis of single crystal alpha alumina particles, the method involving heat treatment of a transition alumina or an alumina precursor in an atmosphere containing hydrogen chloride. I find the process disclosed in the Mohri reference is fundamentally different from the one claimed in the present invention in various respects. First, the objective of the Mohri process is to produce alpha-alumina particles having a **low aspect ratio** (0.5-3 as specified in claim 4 and 0.5-2 as shown in the examples), while the present invention is aimed at producing plate-like alpha-alumina particles having a **high aspect ratio**. Second, the Mohri process involves heat treatment in a highly corrosive gas, while the heat treatment in the present invention can be carried out in an ambient atmosphere, dispensing with special equipment and making the process simpler and less expensive. Third, the Mohri process involves milling, which however is a means of separating aggregated alpha alumina crystals **after** the alpha alumina phase has been formed. By

m.m.

contrast, the present invention involves milling to form a dispersion of an aluminium **precursor** compound in a diluent. As stated above, I consider the JP'818 document to be irrelevant to the present invention. As a result, I also consider any combination of JP'818 and the Mohri reference to be similarly irrelevant. Such a combination would just be a process in which alpha alumina particles are formed in a molten salt and they are then milled as suggested by Mohri to break up the aggregated (fused) particles which form. Using the present invention, such milling after the formation of plate-like alpha alumina particles is not needed because there has been no melting to cause aggregation in the first place.

8. I have been provided with a copy of JPA-60-54916 (hereinafter referred to as "the Hisanori reference") which I have read and understood and translated into English in full. The Hisanori reference discloses a process whereby aluminium sulphate is reacted with a metal carbonate to generate a mixture of **amorphous** alumina and a sulphate. This mixture is then heat treated **above the melting point** of the sulphate to produce hexagonal flake alumina.
9. In the section entitled "Details of the Invention", the Hisanori reference clearly states that: "Upon further heating to above 800° C, eta-Al₂O₃ fine particles are subjected to a dissolution-precipitation reaction in a molten (emphasis added) alkali-metal sulphate to form hexagonal flaky alpha-Al₂O₃ particles". I found nothing in Hisanori to contradict this statement. Thus, Hisanori teaches that **plate-like alpha alumina** can only be formed **above** the melting point of the sulfate. In contrast, the present invention relies on heat treatment **below** the melting point of the diluent in order to achieve plate-like alpha alumina particles. As stated above, this difference is very significant in that a process that can be done below the melting point of the diluent is both cheaper and easier to operate which makes the present invention highly advantageous compared to the process described in the Hisanori reference.
10. To further verify my interpretation of the Hisanori reference, some of the specific Examples presented in the Hisanori reference were replicated in our own laboratory in Western Australia as discussed below in paragraphs 11 to 14.



11. Example 1 in the Hisanori reference involves mixing aluminium sulphate with sodium carbonate in order to produce a mixture of amorphous alumina nanoparticles and sodium sulphate. The mixture was then heat treated at 1000°C, which after washing resulted in plate-like alpha alumina particles. The melting point of pure sodium sulfate is 884°C (according to Wikipedia). It is clear to me that Example 1 teaches that alpha alumina can be formed in a molten sodium sulfate. In order to confirm this, the experiment described in Example 1 was reproduced in our laboratory. Reagents were mixed in a Spex mixer mill in the proportions described in Example 1. X-ray diffraction of the resulting mixture revealed the presence of sodium sulphate. The mixture was heat treated at 1000°C in a ceramic crucible. The heat treatment process yielded a single lump of fused material, similar to that described by Hisanori. It is clear to me that the fused material was generated by melting of the sulfate during heat treatment. The resulting product was washed to remove the sulfate. X-ray diffraction measurements confirmed that the resulting material was alpha-alumina.
12. Example 3 describes the use of a similar mixture of aluminium sulfate and sodium carbonate and demonstrates that heat treatment at 700°C results in eta alumina instead of alpha alumina. This example demonstrates that the mixture must be heat treated at 1000°C as described in Example 1 in order to generate alpha alumina.
13. Example 4 describes a mixture of eta alumina and sodium sulfate, heat treated at 1000°C (above the melting point of sodium sulfate) in order to obtain alpha alumina.
14. Example 2 describes a mixture of aluminum sulfate and potassium carbonate, which is subsequently heat treated at 1000°C in order to obtain platy alpha alumina. While Hisanori does not state what compounds are present after the reagents were mixed together, they are supposed to be a mixture of potassium sulfate and an aluminum compound formed through a displacement reaction between aluminum sulfate and potassium carbonate. In addition, some residual aluminum sulfate must be present, since the aluminum sulfate-potassium carbonate ratio in the starting mixture is 1:2.5 rather than the stoichiometric 1:3. To verify what phases are formed after mixing, the experiment described in Example 2 was reproduced in our laboratory. The reagents were mixed in a Spex mixer mill in the proportions described by Hisanori. X-ray diffraction of the resulting mixture revealed the presence of peaks corresponding to potassium sulfate, plus

a number of minor peaks, which could not be identified because of the weakness of the peaks. While the melting point of potassium sulfate is 1067°C, eutectic mixing with another compound always lower the melting point. (As mentioned above, the off-stoichiometric composition of the starting mixture is likely to result in residual aluminum sulfate.) The resulting mixture was heat treated at 1000°C which resulted in a single fused lump of material similar to that described by Hisanori, clearly indicating that some compounds in the mixture had in fact melted during heat treatment.

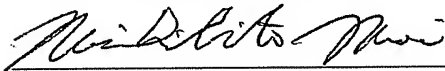
15. Having replicated the experiments described in Examples 1 to 4 and reviewed the full text of the specification, it is clear to me that the Hisanori reference does not teach that plate-like alpha alumina particles can be formed below the melting point of the second-phase compound (sulfate) as claimed in the present invention. Hisanori teaches the opposite – it teaches that the temperature must be raised above the melting point of the diluent to produce plate-like alpha alumina particles. I therefore consider that the Hisanori document is irrelevant to the present invention.
16. I would also consider any combination of the Hisanori reference and the Mohri reference to be irrelevant for the same reasons as those set out above in paragraph 7.
17. I have been provided with a copy of JP 06-329412 (hereinafter referred to as “the Isao reference”) which I have read and understood in full. The Isao reference discloses a method of producing alumina particles having a size of 1-10 microns and an aspect ratio of 1-3, the method consisting of steps of (i) heating aluminium hydroxide in the presence of a fluoride so that conversion to alpha alumina is 10-100% completed and (ii) calcining the resultant alumina at a temperature in the range of 800 – 1500°C in the presence of a boron compound. The process described in the Isao reference is aimed at producing alumina particles having a low aspect ratio compared with the high aspect ratio of the particles of the present invention. With no diluent used, the process described in the Isao reference is fundamentally different from the present invention and for this reason, I consider this reference to be completely irrelevant.



M.M.

18. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,



Michihito Mori

Date: 5/01/09


Witness: Dr Marguerite Port

Date: 5/01/09